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HORSETAIL PLANT PHYTOREMEDIATION POTENTIAL IN THE DECREASE OF HEAVY METAL IRON (Fe) IN LEACHATE AT CILOWONG'S LANDFILL AREA SERANG CITY

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ABSTRACT

Aim: This study aims to determine the potency of horsetail plant to remove heavy metal concentration. **Methodology and Results:** Data was obtained from fifty-four (54) horsetails which were divided into three batches of 12, 18, and 24 plants respectively. Furthermore, the experimental method with the subsurface flow system treatment was used with two samplings for seven days. The results showed that the first, second and third batches gave a 6.83%, 10.28%, and 16.26% impact on reducing concentration of Fe respectively with an approximate average of 11%. Therefore, the detention time of the leachate constructed wetland reactors significantly reduced the Fe contaminant. **Conclusion, significance, and impact of study:** This research provides a solution to mitigate the environment by adopting the accumulator of Horsetail plants (*Equisetem hyemale*) in the constructed wetlands reactor experiment. From the three reactors approximately 0.03 mg/L of heavy metal iron was removed after a seven day run. Therefore, horsetail plant showed a positive impact in reducing environmental pollution, a trend adoptable in wastewater and leachate treatment technology in Indonesia.

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- Heavy metal Fe
- Leachate treatment
- Municipal landfill area
- Phytoremediation
- Potency of horsetail

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1. INTRODUCTION

Serang City has coordinate -48M 627247.70888708 that is determined as a medium-sized with a total population of 655.000 in 2016. The average population growth of the city in the last six years is quite high at an average of 2.0% per year. In 2017 the population density was 2,499 residents per km². This igure is directly proportional to the increase in waste produced by residents estimated at 220 kg/capita per day (Pedoman Inventarisasi GRK-KLH, 2012). In 2018, the waste of the city was 243.372 m3 per day (DLHK, 2018). Only about 34.9% of the waste is transported to landfills, 49.5% is burnt openly, 5.9% not collected, 7.8% throw to river, 1,5% buried and 0.4% composted (Riskesdas, 2018).

The problem of municipal waste at Cilowong's landfill area of Serang City is a complex issue to overcome since it introduces pollution of the surrounding environmental components. Currently, the landfill area collects garbage from the city and the district which are transported by trucks at an average capacity of 5 - 6.5 m³ per truck with an average arrival rate of 70 - 100 trucks daily. The landfill area has a biological leachate pond with an area of 600 m² and a volume of 840 m³ which leachate flows into the river and rice field after sedimentation.

Therefore, it is very important to conduct an in-depth study of the concentration of heavy metals in the leachate, since it has the potential to endanger human health (Abolayo and Tope, T., 2019). According to Article 28 of the 1945 Constitution of the Republic of Indonesia and the regional regulation of Serang City, good environment is the right of every citizen. It stipulates that waste management must preserve environmental functions and public health. But unfortunately, the city waste management system has not met the aim of waste management.

According to Miller and Spoolman (2015) the law of conservation of matter states no atom or is created or destroyed on physical or chemical changes. Therefore, the restoration of land and water contaminated with heavy metals must be carried out immediately through a remediation process, as mandated in Article 54 of the Republic of Indonesia Law 2019 on Environmental Protection and Management.

Therefore, researchers are interested in conducting an experimental study of leachate treatment from the landfill area in Taktakan District, Serang City using phytoremediation techniques of Horsetail or *Equisetum hyemale*. According to Kurniati *et al.*, (2014) lead and chromium contaminant can be removed from landfill leachate using the horsetail plant

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effectively. The use of phytoremediation techniques with some aquatic plants degrades heavy metal content (hyperaccumulator) in their biomass.

This research focuses on reducing the content of heavy metal pollutant Fe in leachate by applying the phytoremediation concept of Horsetail in constructed wetland media. Several tests were carried out from the initial environmental baseline observations at the landfill area suspected of potentially endangering the environment such as Fe, Pb, Cr, and Cd. The heavy metal parameter with the highest concentration and above the environmental quality standard was taken. The concept of phytoremediation of Horsetail in leachate management was used to ensure natural resources in improve the aesthetics of the landfill area.

The results of the study are expected to explain the potential effectiveness of these plants on the potential reduction of heavy metals Fe and reveal the average contribution ability of one Horsetail in removing heavy metal Fe. Based on the problem above, it is necessary to analyze leachate treatment using phytoremediation process of Horsetail to reduce heavy metals Fe. The aims of the research:

1. To analyze the initial environmental baseline conditions for heavy metal concentration in Lead (Pb), Chromium (Cr), Cadmium (Cd), and Ferro (Fe) in the leachate at the Cilowong's landfill area.
2. To measure the potential effectiveness of the Equisetum hyemale in reducing the content of heavy metal Fe in leachate in the settling stream reactor.
3. To measure the contribution of one Horsetail Plant in reducing the concentration of heavy metal Fe in the wetlands construction media.

Hyperaccumulator plants concentrates heavy metals in the biomass at extremely high levels such as aquatic plants. Absorption and accumulation of heavy metals by plants is divided into 3 (three) processes including the absorption of metals by the root, metal translocation from roots to other plant parts, and localization of metals in certain cell parts to maintain and allow metabolism of plants (Irhamni, 2017). The phytoremediation technology can be applied for the recovery of soil, groundwater, and surface water pollution at the landfill area. The specific problem of heavy metals is the dangerous and toxicity imposed on living things upon accumulation through cycles of food chains, intermediaries, or water contaminated by heavy metals.

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Phytoremediation is a process widely used in wastewater treatment technology since it is effective in removing heavy metal. It is rapidly growing due to the underlying advantages, such as being relatively inexpensive compared to conventional methods saving about 75 - 85%. It is a process where certain plants work together with microorganisms in the media (soil, coral, and water) changing contaminants (pollutants) to be less or not dangerous. The method was one of the potential choices for leachate remediation (Haarstad & Maehlum, 1999) and since the mid-1970s had been investigated over a range of scales from ground pond laboratory experiments to full-scale field trials (Jones et al., 2006). Table 1 shows the various type of phytoremediation mechanisms that can treat several chemical compounds. (Susarla, 2002).

In general, plants used in phytoremediation systems have fibrous roots. According to Tangahu and Warmadewanthi (2011), aquatic plants has about ten adventitious roots and 600 lateral roots each that serve as filters for contaminants in liquid waste. Water plants have intercellular spaces or airway holes called *aerenchyma* for transporting oxygen from the atmosphere to the roots (Tangahu & Warmadewanthi, 2011). In wetlands plants, gas transportation occurs by diffusion through channels formed by *aerenchyma*. Carbon dioxide moves in the opposite direction, from the root to the top of the plant. It is a byproduct of transpiration released into the atmosphere through stomata (Cronk, 2001).

Table 1 Phytoremediation mechanism

Type	Chemicals Treated
Phytoaccumulation/Phytoextraction	Cadmium, chromium, lead, nickel, zinc, and other heavy metals, selenium, radionuclides; BTEX (benzene ethyl benzene, toluene and xylenes), pentachlorophenol, short-chained aliphatic compounds, and other organic compounds
Phytodegradation/phytotransformation	Munitions (DNT, HMX, nitrobenzene, nitroethane, nitromethane, nitrotoluene, picric acid, RDX, TNT), atrazine; chlorinated solvents (chloroform, carbon tetrachloride, hexachloroethane, tetrachloroethane, trichloroethane, dichloroethane, vinyl chloride, trichloroethanol, dichloroethanol, DDT; dichloroethane; methyl bromide; tetrabromoethane; tetrachloroethane; other chlorine and phosphorous based pesticides; polychlorinated biphenols, other phenols, and nitriles.
Phytostabilization	Proven for heavy metals in mine tailings ponds and expected for phenols and chlorinated solvents (tetrachloromethane and tri chloromethane)
Phytostimulation	Polycyclicaromatic hydrocarbons; BTEX (benzene, ethylbenzene, toluene, and xylenes); other petroleum hydrocarbon; atrazine; alachlor; polychlorinated biphenyl (PCB); tetrachloroethane, trichloroethane and other organic compounds.
Phytovolatilization	Chlorinated solvents (tetrachloroethane, trichloromethane and tetrachloromethane); mercury and selenium.
Rhizofiltration	Heavy metals, organic chemicals; and radionuclides.

Source: Susarla et al., (2002)

2. RESEARCH METHODOLOGY

Research methodology shows in the cause and effect diagram below:

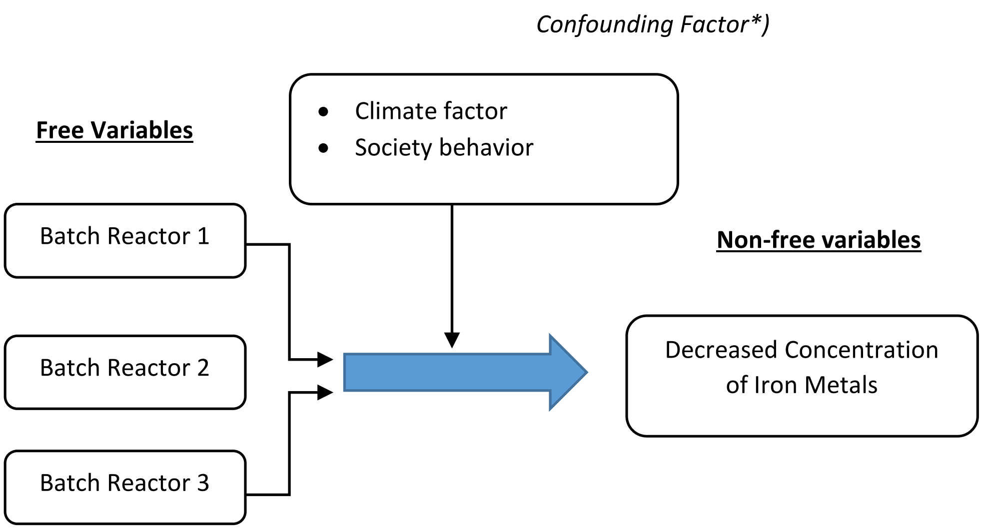

Figure 1 Diagram of cause and effect of research

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The research is conducted at the Cilowong's landfill area, which is located in Panggungjati Village, Taktakan District. The leachate used in the research is obtained from a pond in the landfill area. The location is suitable for research due to the availability of necessary supporting aspects allowing it to run from February to December 2019.

The research uses an experimental method or laboratory testing with phytoremediation techniques of aquatic plants (Figure 1). This experiment was conducted to compare the results on the quality parameters of heavy metal Fe from the three reactor ponds with three variations in the amount of Horsetail plant. A total of 54 clumps of Horsetail are used where 12, 18, and 24 are in the first, second, and third reactor pond respectively.

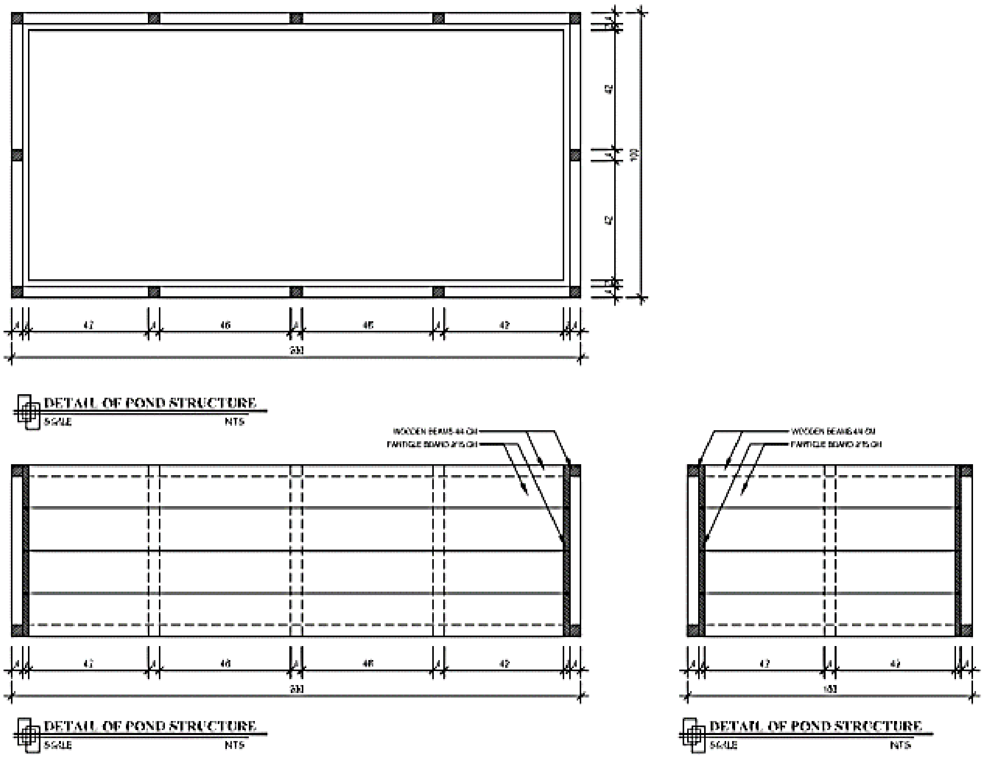

Figure 2 Constructed wetlands reactor design

From Figure 2, the dimensions of constructed wetlands structure are 200 cm length, 100 cm width, and 60 cm height with the mud height of 30 cm, leachate height of 15 cm from the mud and free space height is 15 cm. In this process of making constructed wetlands particleboard, wooden beams, and a tarp are used. The subsurface flow technique is used in the

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constructed wetlands, where the outlet of the leachate is 30 cm from the mud layer base, which flows vertically and moves horizontally. The sample of leachate was taken from an outlet pipe with the tap connecting with the pipe as in picture 3 and picture 4 is the real design.

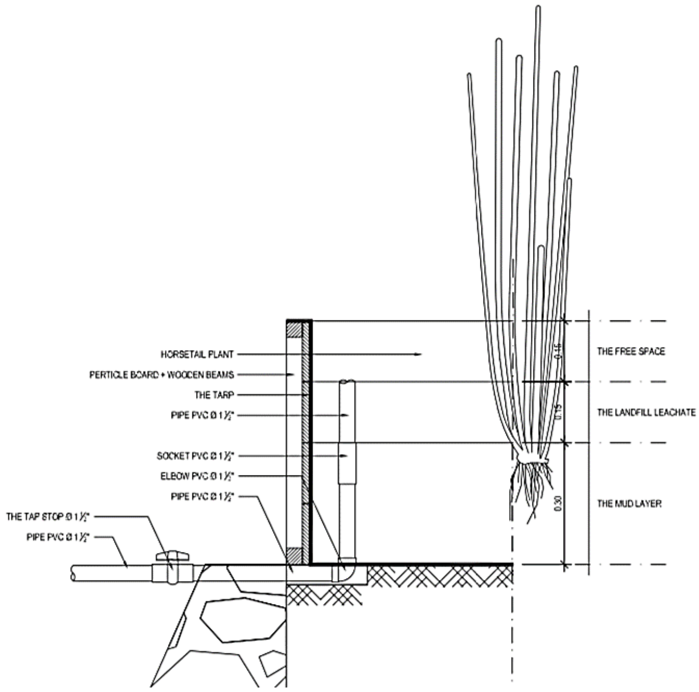

Figure 3 Design of constructed wetlands reactor from side view

Figure 4 Constructed wetlands reactor for phytoremediation of horsetail plants

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Leachate samples used were obtained from Cilowong's landfill area of Serang City which took about 1,200 liters for distribution into three ponds (reservoirs) with different variations of plants. It focused on the effluent of wastewater that enters into the biological leachate pond in the landfill area.

Furthermore, the leachate is placed into gallons of 1,000 liters and 200 liters and taken to the reactor pond made of wood and rafters as per the above mentioned specifications. The reactor that covered by a thick tarpaulin that accommodates mud and leachate. The mud is taken from the muddy rice fields in the surrounding area with a height of 30 cm from the pond bottom.

The sample testing conducted in the laboratory include the testing of existing heavy metal in the leachate pond at the Cilowong's landfill area consisting of Lead (Pb), Chromium (Cr), Cadmium (Cd) and Iron (Fe) in approximately 36 samples at 9 points. After the phytoremediation treatment, a heavy metal content testing is also carried out with a focus on testing the heavy metal Fe content. The result of the analysis of the first test are known to have levels that exceed the specified wastewater quality standards. Final sample testing was conducted on a miniature of leachate pond with phytoremediation technique of Horsetail on muddy soil using an AAS (Atomic Absorption Spectrophotometer) SNI (Indonesia National Standard) 6989.4.2009 whose accuracy and validity can be trusted.

3. RESULTS AND DISCUSSIONS

In the initial environmental baseline, several heavy metal parameters, such as Lead (Pb), Chromium (Cr), Cadmium (Cd), and Iron (Fe), were tested referring to Ministry of Environment and Forestry Regulation Number 5 of 2014 on Wastewater Quality Standards for businesses and/or activities without Wastewater Quality Standards.

Table 2 Wastewater quality standards for measured parameters (examined)

Parameters	Unit	Sample Group I	Sample Group II
Dissolved Iron (Fe)	mg/L	5	10
Lead (Pb)	mg/L	0.1	1
Total of Chromium (Cr)	mg/L	0.5	1
Cadmium (Cd)	mg/L	0.05	0.1

Source: Ministry of Environment and Forestry Standard, 2014

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The results of heavy metal testing on environmental baseline conditions of the heavy metals Fe, Pb, Cr, and Cd studied are as follows:

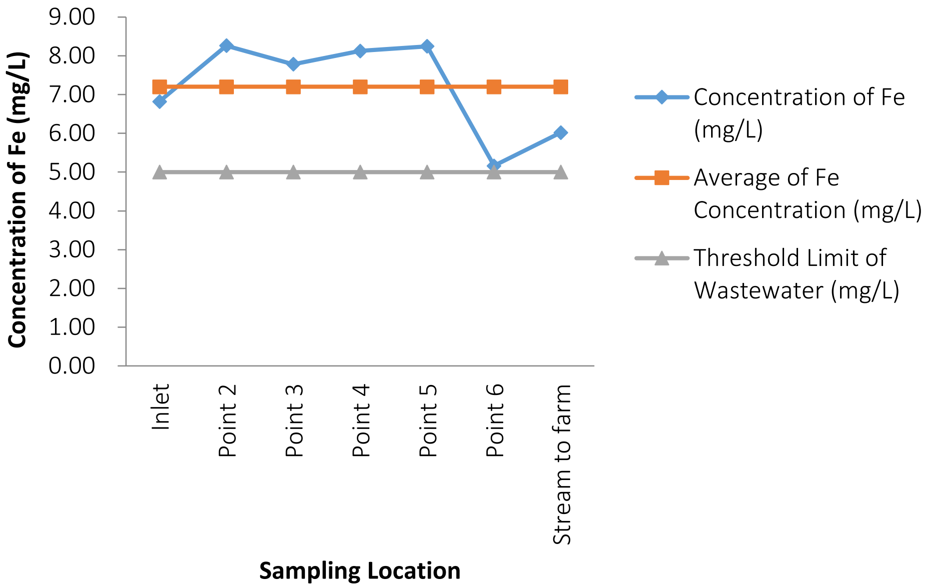

Figure 5 The existing Fe heavy metal parameter test results

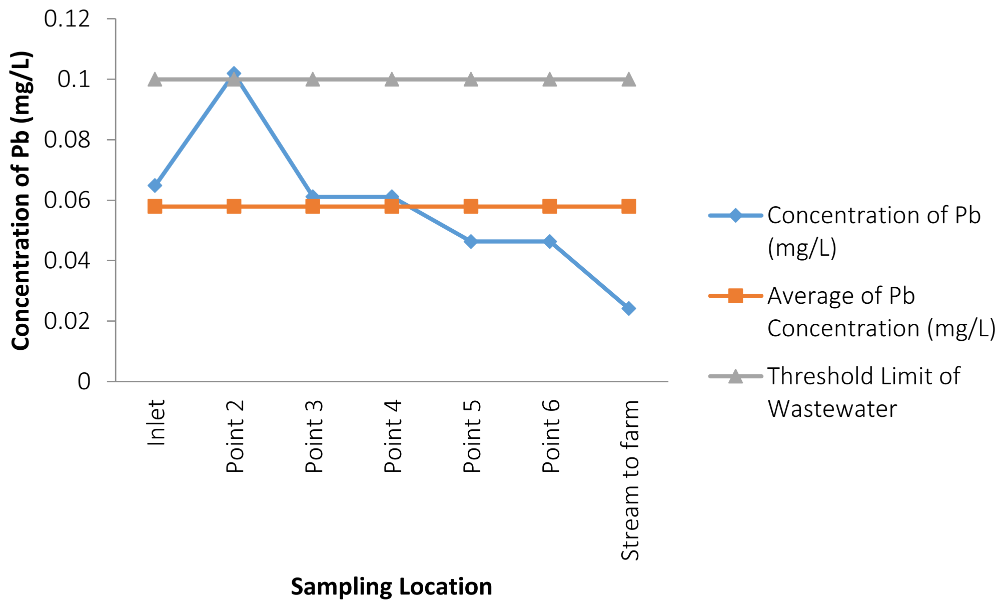

Figure 6 The existing Pb heavy metal parameter test results

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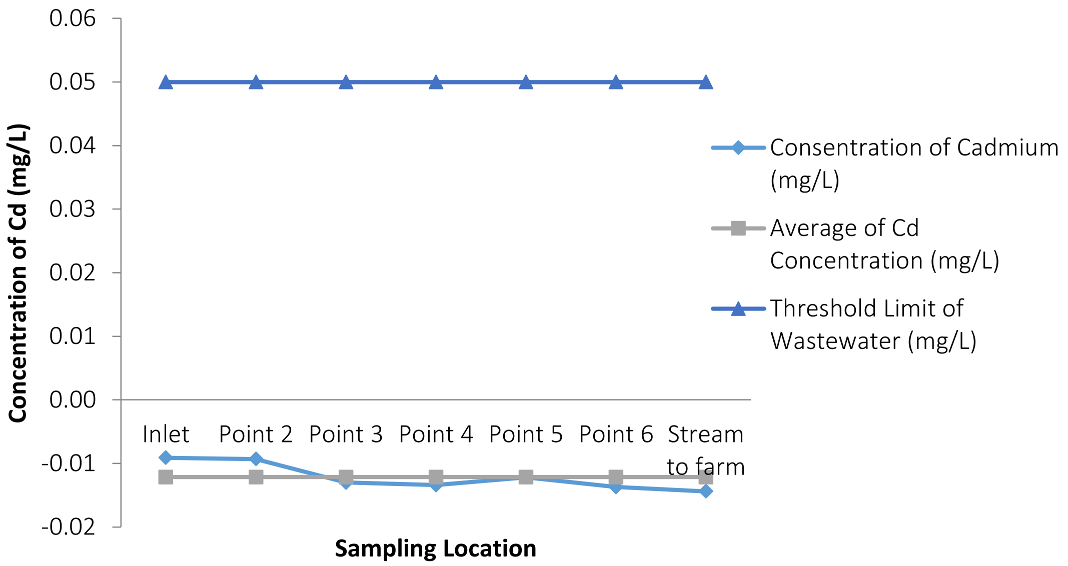

Figure 7 The existing Cd heavy metal parameter test results

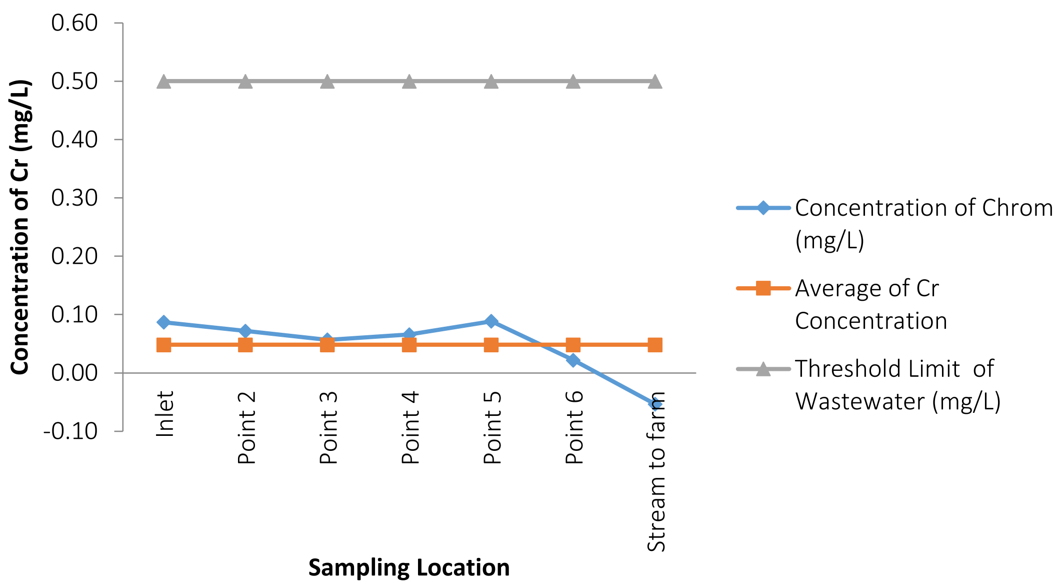

Figure 8 The existing Cr heavy metal parameter test results

Based on the four heavy metal parameters measured above, the condition of the leachate pond of Cilowong's landfill area is declared to be polluted/contaminated with heavy metal Fe

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content exceeding the wastewater quality standard. The concentration of Pb, Cd, and Cr are below the leachate quality standard and wastewater quality standard. A summary of the results of testing heavy metal parameters is presented in Figure 9. Furthermore, the research focuses on the parameters of heavy metals that are above the quality standard, such as Fe, Pb, Cd, and Cr at an average of 7.2019 mg/L, 0.0579 mg/L, +/- 0.01221 mg/L, and 0.048 mg/L respectively.

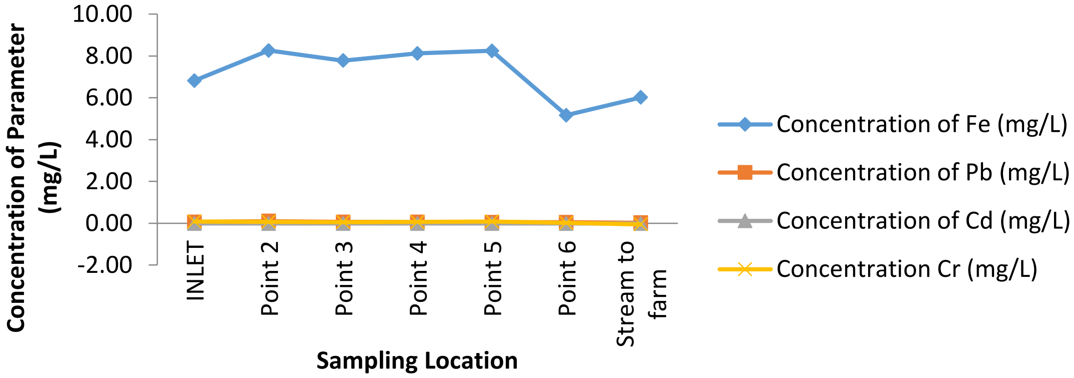

Figure 9 Summary of testing results for measured heavy metal parameters

The results from Figure 9 explained that the Iron concentration was above the quality standard, hence polluted having potential hazardous effect to the surrounding environment, especially to the farmland and soil water. Furthermore, the study continued to experimental design with planting a horsetail in each reactor wetlands pond.

Figure 10 described the result of the concentration of Iron in each reactor wetland where the reactor wetlands 3 have lower average concentration than reactor wetland 1 and reactor wetland 2. Therefore, reactor wetland 3 performed better than others, although the scatter graph showed the dynamic trends.

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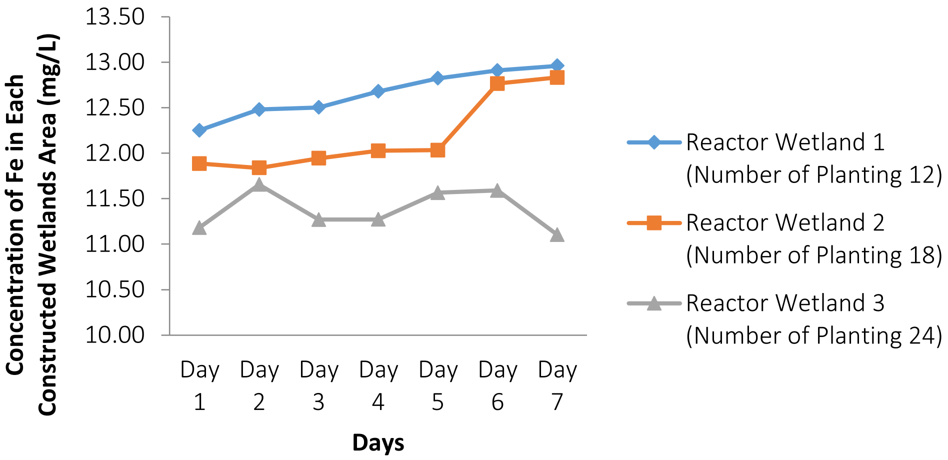

Figure 10 The concentration of Iron (Fe) in each reactor wetlands

Based on Table 3, the effectiveness of plants was influenced by the decrease in Fe concentration of leachate. The highest efficiency value was shown by the results of the sample test on the seventh day of the third reactor pond, which represented the largest population of Horsetail. Therefore, the Horsetail proved to be used as phyatoaccumulators of Iron (Fe) concentrations. The difference in plants in the reactor treatment ponds showed a reduction of Fe leachate by 16.26%. Whereas in the first pond with the least planting results in the lowest efficiency of 6.83% decrease in Fe leachate concentration. The most noticeable difference in efficiency values during the phytoremediation treatment was the first and the last day.

Table 3 The efficiency of horsetail plants ability in reducing fe concentration in each constructed wetlands reactor

Sample	Number of Planting 12 (mg/L)	Efficiency (%)	Number of Planting 18 (mg/L)	Efficiency (%)	Number of Planting 24 (mg/L)	Efficiency (%)
H_1	12.25	9.8%	11.89	12.5%	11.18	17.7%
H_2	12.48	8.1%	11.84	12.9%	11.66	14.2%
H_3	12.50	8.0%	11.94	12.1%	11.27	17.0%
H_4	12.68	6.7%	12.03	11.5%	11.27	17.0%
H_5	12.82	5.6%	12.03	11.4%	11.57	14.9%
H_6	12.91	5.0%	12.77	6.0%	11.59	14.7%
H_7	12.96	4.6%	12.83	5.6%	11.10	18.3%
Average Efficiency		6.83%		10.28%		16.26%

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The efficiency of Fe concentration increased in the reactor pond 1 with 12 Horsetail which is evident in Table 3 and column 3, on the first and seventh day, where the efficiency value were 9,8% and 5% respectively. The condition of reactor pond 2 was the same as reactor pond 1, although there was an increase in average efficiency. Also, to increase the efficiency as evidenced in the Table, analysis of the problems in pond 2 were not different from pond 1. Several signs are attributed to increased effectiveness and ability of the Horsetail as a phytoumulator:

a. Almost 50% of the Horsetail experienced damage and drought on the stem.
b. The condition and characteristic of leachate did not change in color or scent.
c. The volume of leachate in the reactor pond 1 and 2 was sufficient since it covered the plant.

Figure 11 explains the efficiency that occurred in Horsetail upon contact with leachate which showed a decrease in the ability and absorption of heavy metal. The influence of plants on the highest Fe concentration decreased a reactor wetland 3. The resulting 16% efficiency was greater than that generated by reactor wetland 1 and reactor wetland 2. From Figure 11, the number of plants used as leachate phytocumulators, the greater the decrease in Fe concentration. In other words, the higher the phytocumulators, the less the concentration of a metal in leachate. The changes that occurred in the reactor wetland 3 were the reduction of leachate discharge and physical characteristics and conditions. Discoloration and reduction of discharge volume in the leachate emerged moss in the mud due to the phytoremediation process carried out by Horsetail.

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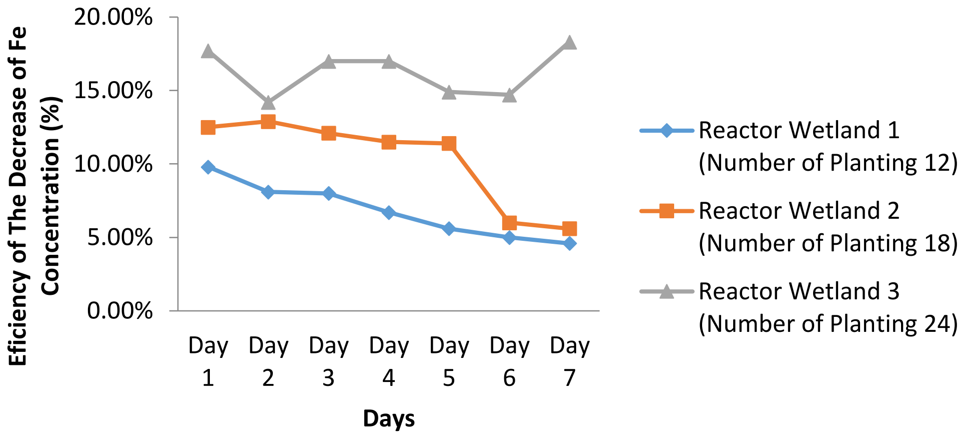

Figure 11 Efficiency graph for the number of plants to decrease Fe concentration in each reactor wetlands

From this research, Horsetail influences the decrease of Fe concentration in the pond after treatment which is indicated in the variation of plants and Fe content in each treatment. Therefore, the Horsetail affects the increase of Fe content in the roots and stems. There is a relationship between the decrease in Fe concentration in the planting media and its absorption in Horsetail. Therefore, a decrease in Fe concentration in the planting media results to an increase in the Horsetail.

The ability of Horsetail to absorb and accumulate heavy metals is evident from the large concentration of iron (Fe) left in leachate. Based on the testing results of the concentration of iron (Fe) in leachate (Table 3 and Figure 11), the results of the sample of leachate from the third pond containing most Horsetail. The result in the seventh day showed the largest decrease in iron concentration (Fe) compared to leachate samples from the first and second pond, which filled Horsetail plants with a smaller population. It is estimated that the plant had absorbed some iron (Fe) from leachate.

In the reactor for experiment 1 and 2, the concentration of iron metal (Fe) reduced on a lower margin than the third pond. It was also marked by the growth of moss in the mud that became food for survival resulting to a decrease in the concentration of iron (Fe). Comparison of test results showed that the concentration of iron (Fe) in the existing pond at the landfill area was significantly different from the reactor pond with Horsetail. This suggested that the plant tested on constructed wetlands could affect the reduction of iron (Fe) concentration in leachate

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in real conditions. The efficiency of plants on decreasing Fe concentration of leachate can be seen in Table 3.

In Table 4 below, the average efficiency is obtained through phytoremediation to decrease the concentration of Fe in leachate.

Table 4 The efficiency of planting in phytoremediation reservoir to decrease Fe concentration in leachate

Pond of Phytoremediation Process	Number of Planting	Average of Concentration Fe (mg/L)	Efficiency Average (%)
Pond 1	12	12.66	6.83
Pond 2	18	12.19	10.28
Pond 3	24	11.38	16.26
	54	12.08	11.38

Based on Table 4 above, the average Fe concentration of leachate in the phytoremediation treatment for the reactor pond 1 and 2 showed little variation but in terms of effectiveness, pond 2 is higher than pond 1. The difference in efficiency values was strongly influenced by the value of the Fe concentration test results from the first to the seventh day. The results of testing leachate in pond 1 and 2 from the first day to the seventh day showed no changes. The average amount of Fe concentration for pond 3 was 11.38 mg/L with the highest decreasing value among the other two ponds at 16.26% signifying action and reaction in the Horsetail and leachate.

The major difference between pond 1 and 2 was the acquisition of Fe content from the first day to the seventh day. However, there was a decrease from the graph of the test results in the pond 1. The total efficiency using three variations of the 54 Horsetail reduced Fe contaminants in leachate by 11.38%. The accumulation of a reduction in the amount of iron (Fe) concentration from the initial test results before being treated in leachate was 13.59 mg/L - 12.08 mg/L = 15,067 mg/L. The average for each Horsetail from the three phytoremediation ponds can only reduce Fe concentrations in leachate by 1.5067/54 = 0.03 mg/L per Horsetail for seven days of the trial period.

The ability of Horsetail to absorb and accumulate heavy metals is evident from the large concentration of iron (Fe) left in leachate. Based on the test data on the concentration of iron (Fe) in leachate, water samples from pond 3 with a large Horsetail population (24 clumps)

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showed a smaller concentration of iron (F) than the sample water from pond 1 and 2 with few Horsetail (12 and 24 clumps respectively). It is estimated that the plant has absorbed some iron (Fe) from leachate.

The problems of the phytoremediation using Horsetail were the iron (Fe) concentration in leachate which tends to increase during the observation period and there was a difference in the decrease of the average yield on each pond. Therefore, the following assumptions cause an increase in the concentration of iron (Fe) metal in the three prototypes which need further investigation:

1. The number of Horsetail in the three ponds were too small to optimally reduce the metal content of iron (Fe) in the leachate.
2. The leachate stay time for Horsetail is long due to the absence of its circulation and flow processes in the existing reservoir at the Cilowong's landfill area.
3. Absence of acclimation process on Horsetail meant no adaptations to the new environment and conditions.
4. Unsterilized mud used in the pond was suspected to contain a very high metal iron (Fe).

From the assumption mentioned above, it can be concluded that the mud used in the construction of wetlands taken from the rice field contains high content of iron (Fe) metal. This is based on the environmental and regional conditions as well as the characteristics and types of soil. In general, the type of soil is Alfisol that has developed with the characteristics of the soil profile forming a horizon sequence of A/E/ Bt/C which is formed through a combination of podsolization and lateralization in wet climate regions under hardwood forest stands (Tan, 2000). It is soiled in areas that have a high rainfall to move clay down to form an argillic horizon a soil layer formed due to the accumulation of clay. It is a fertile soil with a high base saturation of 50 %. The land is formed under various forests or covered in bushes (Miller and Donahue, 1990) where Fe concentrations are very high. Alfisol soil usually has a high Fe and low humus levels which causes the soil to be red (Wirjodihardjo 1963). The test was conducted on the concentration of iron (Fe) in leachate mixed with mud soil to enhance its content to obtain a value of 13.59 mg/L (base point conditions).

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4. CONCLUSIONS

From the trials that have been described in the previous chapter with variations plants in 3 trial ponds of the application technique of construction wetlands (Constructed Wetlands) with mud flooded with leachate and planted with Horsetail, it can be concluded that phytoremediation has a great potential to be applied to various types of the severe environmental pollutants such as contamination on the surface and subsurface of the land and waters. Those are caused by leachate waste generated from the process of extracting solid waste (rubbish) in the area of the landfill.

The application of Horsetail plants (*Equisetum hyemale*) with the constructed wetlands technique can reduce the content of heavy metals of Fe in leachate up to 20%. In developing cities such as the city of Serang with yearly population increase the application of phytoremediation is important due to the unavailability of optimum landfill area. The application is an effort to reduce pollution in the landfill area of Indonesia.

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